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Magnetic coding in systems of nanomagnetic particles

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Nanomagnetic systems show exotic memory effects in the dc magnetization as a function of temperature. We present magnetization measurements on systems of nanomagnetic particles that show that such systems store the memory of either decrease or increase of magnetic field enabling a magnetic "coding" of "0"s and "1"s. Application of a field larger than a critical field, H^* erases the memory effects. We show that this behavior can be explained by a wide distribution of grain sizes having different blocking temperatures. The effect can be used as a tool for measuring spacial and temporal magnetization changes of a magnetic surface. © *2006 American Institute of Physics*.
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Magnetic nanoparticles are gaining increasing interest due to their vast technological potential as magnetic based nanoelectronic devices.[1–6] One of the exotic phenomena observed in systems of single domain magnetic nano-particles is a history-dependent magnetic memory in the dc magnetization of the system as a function of temperature.[7,9–11] Starting at high temperature, the system is steadily cooled in a small magnetic field, H, and the magnetization, M, measured as a function of temperature. At intermediate temperatures the cooling is arrested and the field switched off for a few hours before being restored. When the system is heated from the lowest temperature, $M(T)$ shows wiggles at all T steps where H was previously switched off, apparently keeping a memory of the temperature arrests.

Two explanations have been suggested for such behavior. The first attributes the history dependent $M(T)$ to aging and concomitant memory effects in a spin glass phase.[7] This explanation hinges on the frustration of the magnetic moments of the particles due to dipole-dipole interactions, giving rise to deep energy valleys trapping the system for extended times.[8]

An alternative origin for the memory effects is based on a broad distribution of particle size.[9–11] The polydispersity of the particle volumes leads to a wide distribution of blocking temperatures, T_B. The memory effects are a consequence of the fact that the system is arrested at temperatures which lie between blocking temperatures of different sized particles.

In this letter, we present an experimental study of nanomagnetic systems and show that the effect of a sudden *increase* of magnetic field can also be stored in the system as long as the field is not too high. Using numerical simulations we show that this behavior is consistent with a wide distribution of particle sizes having different blocking temperatures and discuss possible applications of this effect.

The results presented in this letter were obtained on systems of $Ni_{0.35}Zn_{0.65}Fe_2O_4$, however similar results were obtained for systems of $NiFe_2O_4$ particles embedded in a SiO_2 matrix[9] and on Ni particles embedded in a Au matrix (preparation described in Ref. 12). The $Ni_{0.35}Zn_{0.65}Fe_2O_4$ particles were prepared by mechanical alloying processes utilizing Fritsch Planetary Mono Mill Pulverisette 6. The mean crystalline size estimated from powder x-ray diffraction analysis was 27 nm. Figure 1 depicts a transmission electron microscopy (TEM) micrograph of such a sample, exhibiting particles with sizes ranging from 10 to 100 nm.

Our experiments were carried out in accordance with the following cooling and heating protocol. At $T=300$ K a mag-

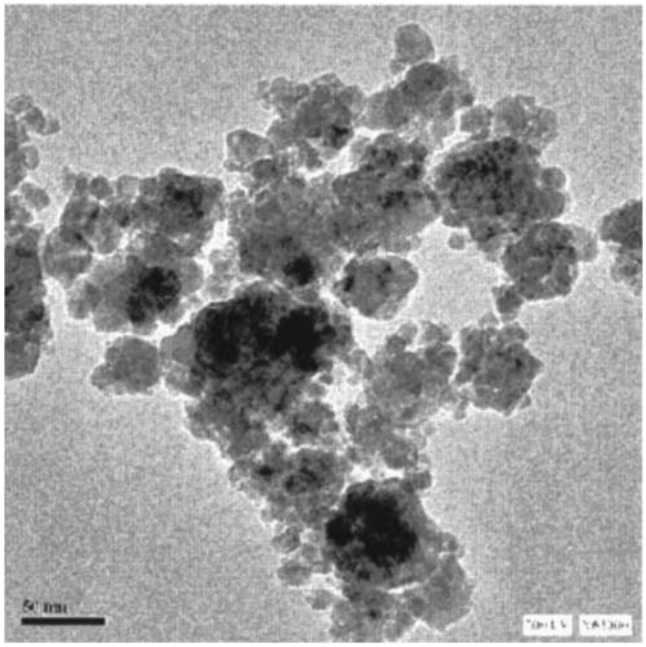

FIG. 1. TEM micrographs of $Ni_{0.35}Zn_{0.65}Fe_2O_4$ particles with diameters ranging from 10 to 100 nm.

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netic field of $H=50$ Oe was applied and the magnetization (M) measured in a superconducting quantum interference device. Keeping the field on, the temperature (T) was lowered continuously at a rate of 2 K/min to $T_1<300$ K and then the field was either switch off or increased to 400 Oe and arrested at this state for 4 h before a 50 Oe field was restored. The sample was than cooled to $T_2<T_1$ after which the field was changed again while arresting the system for 4 h. Finally, the field was changed back to 50 Oe and the sample was cooled to a minimal temperature of 10 K. The system was then heated back to room temperature in the presence of $H=50$ Oe and the heating profile of $M(T)$ was monitored. Results for different changes of magnetic field are summarized in Fig. 2. It is seen that the heating $M(T)$ curve detects both decreasing and increasing magnetic field changes which occur during the cooling protocol. Hence, it is possible to code binary numbers like 0 and 1 by defining the "H decrease" as 0 and "H increase" as 1. These can be decoded by heating the system at a constant rate in presence of a constant magnetic field.

The ability to store the information of many changes is restricted to small magnetic fields. If a field above a critical value, H^* is applied, it erases the memory effects of the smaller H changes. An example is shown in Fig. 3 where applying a magnetic field of 800 Oe results in erasing the fingerprint of a previous magnetic field change.

The observed behavior is naturally understood on the basis of the wide size distribution of particles. Due to the polydispersity, the system contains a wide distribution blocking temperatures, T_B. Thus, for each arrested temperature some of the particles are superparamagnetic while others are blocked. An increase or decrease of magnetic field will be imprinted in large grains but not in small ones since upon restoring the field (after arrestment), the small particles will show facile response while the large ones will not. Further cooling of the sample causes the magnetization in the small particles to increase with temperature while M in the large one will remain almost constant. As T is increased again, M for small particles decreases while for large particles it initially increases before dropping off at $T>T_B$. Due to the wide size distribution, any arresting temperature will correspond to a T_B of some particles, hence, the $M(T)$ during heating will show wiggles at the cooling arrested temperatures, thus mimicking the cooling curve and "remembering" the arrested temperatures. This explanation is not related to complex spin-glass type interactions and applies for noninteracting single-domain magnetic particles.

The earlier reasoning applies only if the H is not too large. If the applied field is large enough to flip all (or most) of the particles, it will erase the previous imprinted memory. In order to understand this we consider, for simplicity a system of magnetic nanoparticles having three distinct volumes $V_1<V_2<V_3$ corresponding to three blocking temperatures $T_{B1}<T_{B2}<T_{B3}$. During the cooling process the system is arrested at two temperatures, T_1 and T_2 so that $T_{B1}<T_1<T_{B2}<T_2<T_{B3}$. Arresting the system at T_2 and changing the magnetic field causes the signature of the change to be imprinted only in the largest particles, V_3. Further cooling the sample does not cause magnetic changes in these particles which are blocked in this temperature range, neither does a mild magnetic field change at T_1 which will give rise to a memory sign imprinted in V_2 particles. However, if at T_1, the applied magnetic field is large enough to flip the spins of V_3 that were blocked at T_2, the magnetic state of the large particles will change even at temperatures well below their blocking temperature. In such a case, the memory imprinted at T_2, is erased.

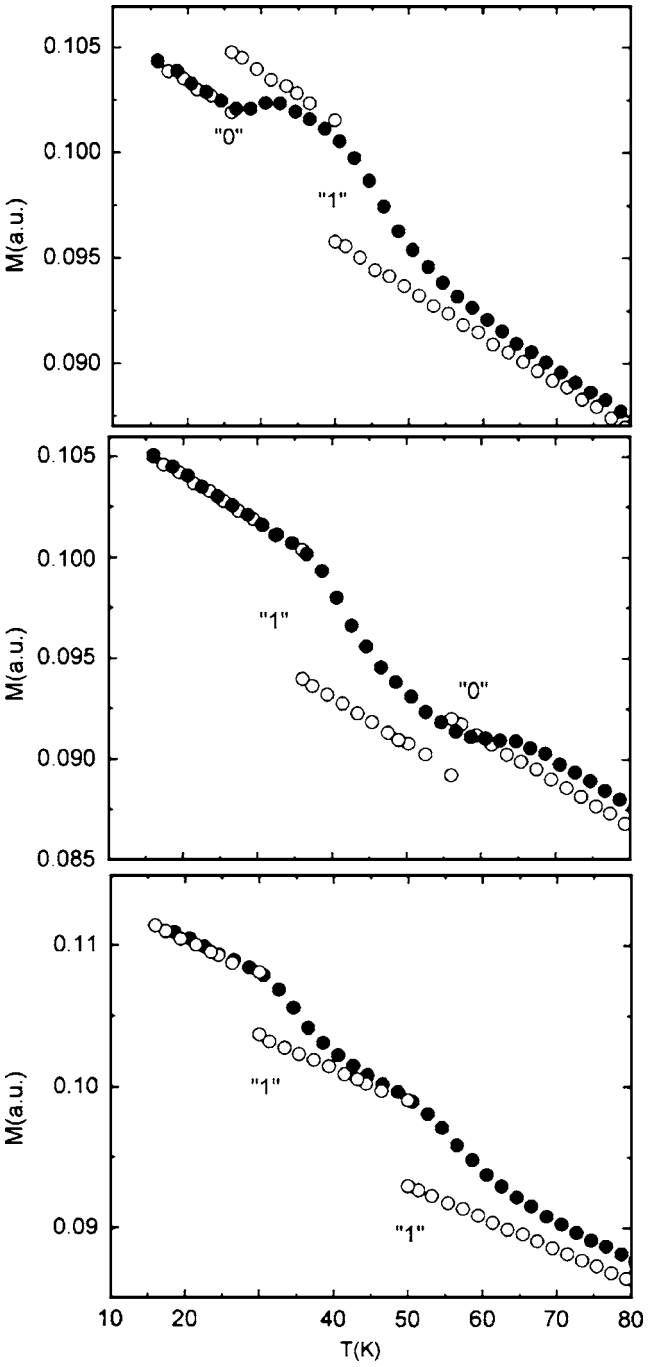

FIG. 2. $M(T)$ curves while cooling (open circle) and heating (solid circle) of the nanomagnetic particle systems at 50 Oe. During the cooling process the following magnetic fields changes were performed: At 40 K a 400 Oe is applied and at 24 K the field is switched off (top panel), at 50 K the field is switched off and at 30 K a 400 Oe field is applied (middle panel), a 400 Oe field is applied both at 50 and at 30 K (bottom panel). Note that the heating curves show different wiggles for the increase or decrease of magnetic field during the cooling process.

In order to simulate the coding curve we considered a system of magnetic nanoparticles having a tetramodal distribution (four distinct particle sizes). We used a two state rate equation assuming the anisotropy energy is large enough than the thermal fluctuation to be in Kramers regime. For simplicity we also assumed that the magnetic field is applied

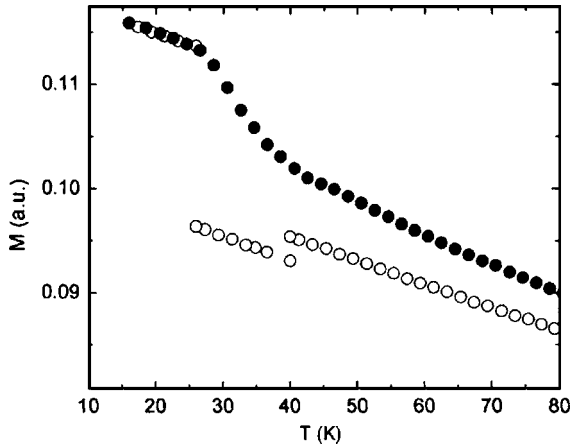

FIG. 3. $M(T)$ curves for the cooling (open circle) and heating cycle (solid circle) while switching off the field at 40 K and applying an 800 Oe field at 24 K. In this case, the signature of the field switching off at 40 K is erased from the heating curve.

along the direction of easy axis of the nanoparticles, so there is no moment rotation but only magnetic orientation flip. Figure 4 shows the results of our simulation for two cases of cooling the sample at a field H, switching the magnetic field off at $T=60$ K and applying a magnetic field larger than H at $T=20$ K. In the first case the applied field is $2H$ while in the second it is $5H$. It is seen that for the mildly applied magnetic field case the heating curve shows magnetization "wiggles" in accordance with the field changes during the cooling process. These are very similar to the experimental results. Applying a large field (bottom panel of Fig. 4), on the other hand, results in the erasing of the signature of the field switch off at 60 K.

The observed effect can be applied to studying the special or temporal variation of magnetic state of a magnetic surface. To study the spacial variation the polydispersed nanomagnetic sample (PDNM) should move over the surface. During its scan its temperature will be decreased at a constant rate. The scan will end at the lowest temperature assigned to the PDNM sample. After the end of the scan the PDNM will be heated at a constant rate to decode the recorded magnetic data as a function of temperature, which in turn will give the spacial distribution of the magnetic state of the surface. The ability to easily produce very small samples can result in very high spacial resolution. A similar algorithm can be used in case of temporal variation, in which the PDNM should be kept fixed at a point and the variation of the magnetic state of the surface as a function of time will be recorded. The sensitivity of the memory device (PDNM) can be increased by increasing the magnetization of the nanomagnets. The resolution can be improved by properly choosing the relaxation times of the nanoparticles and their distribution in the sample.

In conclusion, we have demonstrated that a nanomagnetic particle system can be used to encode a sudden increase or decrease of magnetic field, while cooling the system. The information can be decoded by measuring the $M(H)$ curve during heating. The memory is erased if a large magnetic field is applied. This behavior can be explained as resulting from the wide distribution of particle volumes, without the need of complicated spin-glass type interactions, demonstrating that the lack of particle uniformity can lead to exciting and exotic magnetic effects. The observed effects suggest the possible use of polydispersed magnetic nanoparticles as a "magnetomicroscope" capable of scanning and storing the spacial and temporal magnetic state changes of a magnetic surface.

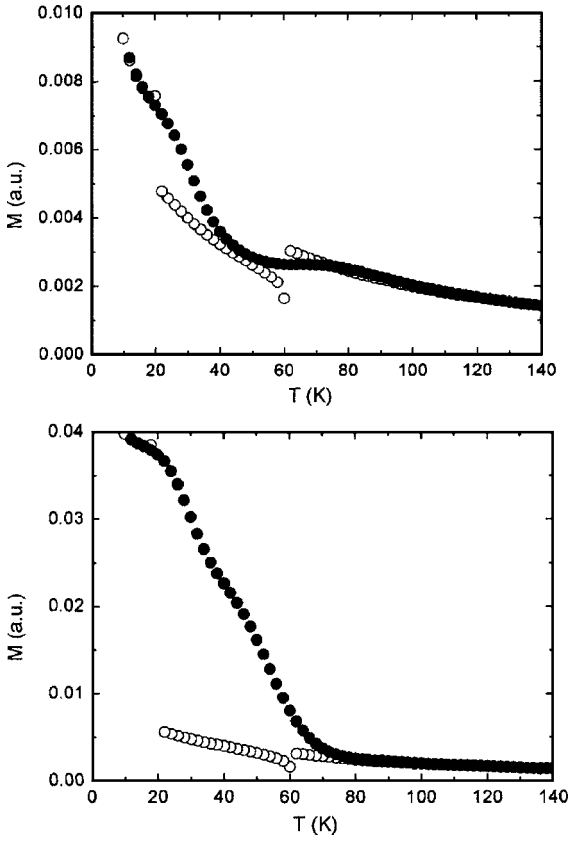

FIG. 4. Results of our simulations for a system in which the sample was cooled (open circle) at a magnetic field, H. At $T=60$ K the field is switched off and the temperature of the system is arrested before restore the field H and at 20 K a field of $2H$ (top panel) or $5H$ (bottom panel) is applied. Note that the application of a large field erases the effect of the previous magnetic field switching off. The solid circles in the figure are the magnetization during heating cycle.

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